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Effect of growth temperature on polytype transition of GaN from zincblende to wurtzite

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Abstract

We have investigated effect of growth temperature on the polytype conversion of cubic GaN (c-GaN) grown on GaAs (001) substrates by MOVPE. It was found that the polytype transition of GaN from zincblende (cubic) to wurtzite (hexagonal) structures is much dependent on the growth temperature. Transmission electron microscopy (TEM) observations demonstrate that the GaN grown layers have the cubic structure (c-GaN) and contain bands of stacking faults (SFs) parallels to {111} planes. For low growth temperatures (~900 °C), XRD results demonstrate that the GaN grown layers with the cubic phase purity higher than 85% were obtained. No different types of single diffraction spots, indicating the incorporation of single-crystal h-GaN, on the selected area diffraction (SAD) pattern was observed. It is also found that a density of SFs decreases with the distance from the interface of c-GaN/GaAs. On the other hand, GaN layers exhibited a transition from cubic to mixed cubic/hexagonal phase under conditions of increasing growth temperature (~960 °C) as determined using TEM-SAD technique with complementary XRD and PL observations. In addition, the optical characteristics of c-GaN layers are shown to be very sensitive to the presence of the single-crystal h-GaN.

Keywords: GaN; Transmission electron microscopy; Growth temperature; Polytype transition; Metalorganic vapor phase epitaxy

1. Introduction

Gallium nitride (GaN) has been intensively investigated as a material for blue-light-emitting diodes (LEDs) and laser diodes (LDs) [1–7] because GaN is the direct wide band gap semiconductor with band gap energy $E_g = 3.39$ eV at room temperature and forms a continuous range of solid solution with AlN and InN, which covers the band gap between 6.2 and 0.7–1.0 eV. It is well known that GaN usually crystallizes in the wurtzite (hexagonal) structure (h-GaN), which is quite different from the other III–V compounds. However, GaN films with the zincblende (cubic) structure (c-GaN) have been grown on GaAs by metalorganic vapor phase epitaxy (MOVPE) [8]. Since then, the MOVPE growth of c-GaN films with higher crystal quality on GaAs (001) and 3C-SiC (001) substrates has been reported [9–14]. One of the most prospective advantages of c-GaN is the growth on cubic substrates, like GaAs, 3C-SiC, Si and MgO, which can be easily cleaved along the substrate facet being good for the fabrication of laser structures. Furthermore, the electronic properties of c-GaN, such as higher mobility, which results from lower phonon scattering in higher crystallographic symmetry [15], should be superior to those of h-GaN. However, the inferior crystal quality with the incorporation of hexagonal phase inclusion obstructs explicating the optical properties of c-GaN. It is known that the crystal quality strongly depends on the growth conditions, particularly the growth temperature, whether the cubic or hexagonal phase is grown even if cubic substrates are used [16].

In this report, the growth of c-GaN is describes from the viewpoint of crystal structure control. The effect of growth temperature in the MOVPE grown c-GaN films has been investigated. It is found that the polytype transition of GaN from cubic to hexagonal phase is much dependent on the growth temerature. Finally, influence of growth temperature on the generation of planar defects (stacking faults and twins) and single phase h-GaN in the c-GaN films is discussed.

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2. Experiments

c-GaN layers were grown on semi-insulating (SI) GaAs (001) substrates at temperature ranging from 900 to 960 °C by metalorganic vapor phase epitaxy (MOVPE), which has been described elsewhere [16,18]. Trimethylgallium (TMGa) and 1,1-dimethylhydrazine (DMHy) were used as the precursors of Ga and N, respectively. c-GaN films were grown using a two-step growth process. An about 100 nm thick GaAs buffer layer was firstly grown at 700 °C, followed a 20 nm thick c-GaN buffer layer at 575 °C with a V/III ratio of 100. Then an about 1.5 μm thick c-GaN film was grown at 900–960 °C with a V/III ratio of 25. The growth rate was 6 μm/h determined by the TMGa molar flow rate of 18 μmol/min.

Structural and optical properties of c-GaN films were investigated by XRD, TEM and photoluminescence (PL), respectively. The TEM specimens were sliced with a diamond cutting saw to a thickness of 0.3 mm in the direction parallel to (1–10) plane. The sliced platelet specimen was mechanically thinned down to 0.01 mm and was finally polished by argon ion milling (GATAN PIPS model 691).

3. Results and discussion

X-ray diffraction (XRD) was primarily used to characterize the structural quality of the MOVPE grown c-GaN films on GaAs (001) substrates at different growth temperatures (900 and 960 °C). It indicates that all c-GaN films condensed in cubic phase. As shown in Fig. 1(a), the diffraction peaks of c-GaN were clearly observed at 39.98°. On the other hand, no diffractions were observed at 34.56° and 36.84° where h-GaN would give the (0002) and the (1−101) diffractions, respectively. Full-width-at-half-maximum (FWHM) of ω-scan for the c-GaN (002) plane was increased from 30.0 arcmin to 40.4 arcmin with increasing growth temperature from 900 °C up to 960 °C. In order to investigate effect of growth temperature on the generation of hexagonal phase in c-GaN films, the volume amount of hexagonal phase inclusion is estimated from the ratio of the integrated XRD intensities of the cubic (002) and hexagonal (1–101) planes measured by ω-scan [7]. It demonstrates that the hexagonal phase inclusion much dependent on the growth temperature. In fact, the cubic phase purity higher than 85% was achieved for the c-GaN film grown at 900 °C. With increasing the growth temperature up to 960 °C, the hexagonal phase inclusion in such high-temperature grown c-GaN film is relatively higher (>40%) than that in c-GaN film grown at 900 °C (<15%).

Fig. 1(b) shows the low-temperature (5.5 K) PL spectra of the c-GaN films grown at temperatures of 900 and 960 °C. For the 900 °C grown layer, the PL spectrum consists of excitonic emission of c-GaN at 3.267 eV with FWHM of 15 meV. The donor–acceptor (D–A) pair recombination around 3.156 eV is very weak. It is also clearly seen in the figure that the other luminescence peaks at around 3.415 and 3.469 eV corresponding to the emission of h-GaN is not observed. It is also noticeable that the orange luminescence observed at 2.05 eV [19] is hardly observed from this c-GaN film. As the growth temperature rises, the hexagonal peak of GaN at 3.469 eV becomes stronger while the cubic peak (3.267 eV) becomes weaker. In addition, the yellow luminescence peak of h-GaN at ~2.2 eV is clearly seen. The difference in the PL spectra is in connection with the different amount of hexagonal phase inclusion in the c-GaN films, which agrees well with the results of XRD.

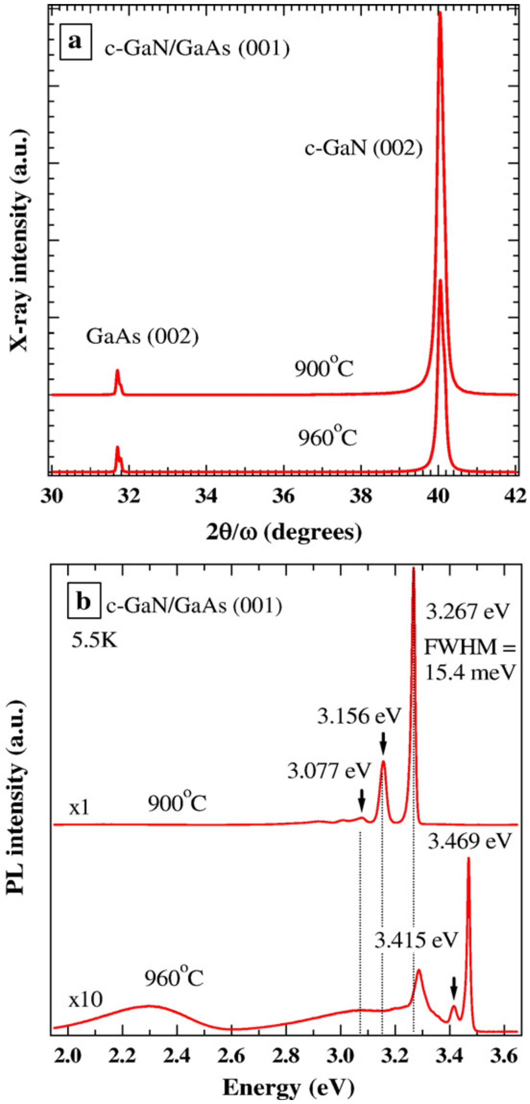

Fig. 1. (a) X-ray diffraction (002) profiles of c-GaN films grown at different growth temperatures of 900 and 960 °C. (b) Low-temperature (5.5 K) PL spectra of the corresponding c-GaN films.

To understand the above results, the c-GaN films were studied by cross-section TEM and selected area diffraction (SAD). The bright-field (BF) TEM micrographs and the SAD patterns in Fig. 2 represent the c-GaN films grown on GaAs (001) substrates at the growth temperatures of 900 °C (Fig. 2(a) and (b)) and 960 °C (Fig. 2(c) and (d)). For lower growth temperatures (~900 °C), the SAD patterns from the interface portion clearly indicates that the GaN grown layer has the cubic structure (Fig. 2(b)). No different types of single diffraction spots on the SAD pattern were observed. However, the microstructure is characterized by a high density of stacking faults